THE COLLISIONAL DE-EXCITATION
OF
ARGON AT VARIOUS PRESSURES

Gustav Henri Stiehl



TEANUATE SCHOOL

United States Naval Postgraduate School



THESIS

THE COLLISIONAL DE-EXCITATION

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by

Gustav Henri Stiehl IV

June 1970

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Argon at Various Pressures

by

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Submitted in partial fulfillment of the requirements for the degree of

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from the

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ABSTRACT

Research grade argon at pressures from 1 to 800 torr was excited by 1.5 MeV protons from a Van De Graaff accelerator. The intensities of the 7635.1 Å and 8115.3 Å spectral lines were analyzed for their dependence on pressure. Theoretical equations were derived for the intensity as a function of pressure which agreed quite well with the experimental data. The collisional de-excitation rate, K_i , for the 7635 Å transition was found to be $3.78 \pm .70 \times 10^{-12} \text{ cm}^3 \text{ sec}^{-1}$ and the collisional de-excitation cross section, σ_d , was found to be $6.7 \pm 1.2 \times 10^{-17} \text{ cm}^2$. For the 8115 Å transition, K_i was found to be $3.81 \pm .53 \times 10^{-12} \text{ cm}^3 \text{ sec}^{-1}$ and σ_d was found to be $6.76 \pm .95 \times 10^{-17} \text{ cm}^2$.



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I. INTRODUCTION

In recent years scientific interest in argon has grown due to its characteristically "ideal" structural make-up. A study of ion-atom collisions and the resultant excitation and de-excitation is of fundamental interest with important applications in many plasma devices and in the physics of the upper atmosphere. In this study the formation of Ar* from the impact of 1.5 MeV protons on research grade argon was investigated. The purpose of the experiment was to study the reaction

Previous works by Thomas and Gilbody [Ref. 1]; Van Eck, et. al. [Ref. 2]; Thomas [Ref. 3]; and Jaecks, et. al. [Ref. 4] studied the resultant excitation caused by protons and helium ions on argon. These studies were made at ion energies from the KeV range up to a maximum of 1.0 MeV and at pressures such that the single hit condition [Ref. 5] was applicable. Primary interest was focused on the ArII spectral lines in the visible range in order to determine excitation cross sections. In this study the pressure was varied from 1 to 800 torr in all cases. The single hit condition does not apply at such high pressures.

The two ArI transitions, $4p\left[\frac{3}{2}\right]^5 - 4s\left[\frac{3}{2}\right]^5$ (Paschen notation: $2p_6 - 1s_5$) at a wavelength of 7635.1 Å and $4p\left[\frac{5}{2}\right]^5 - 4s\left[\frac{3}{2}\right]^5$ ($2p_9 - 1s_5$) at a wavelength of 8115.3 Å (Fig. 1) were examined in detail in this investigation. The experiment was carried out to find the relationship of the spectral line intensity as a function of pressure. From the experimental data the collisional de-excitation rate and collisional de-excitation cross section were determined.



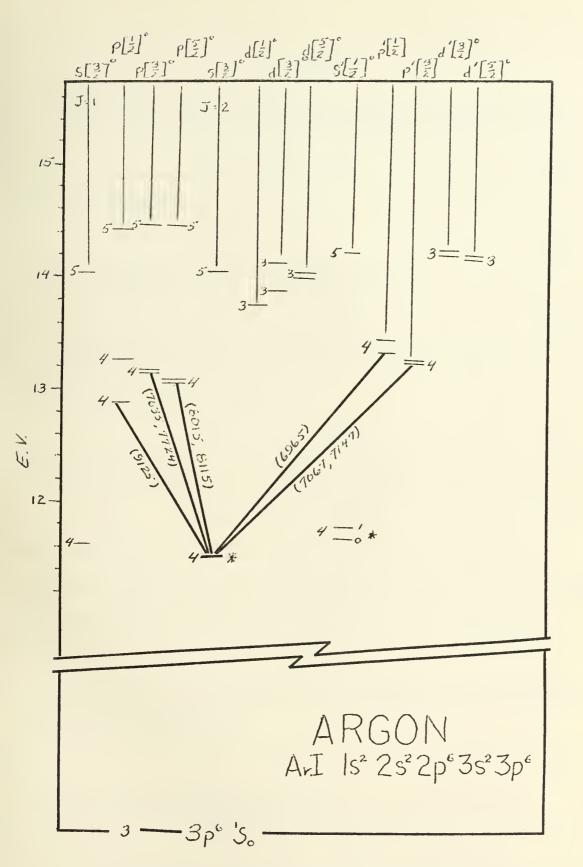


FIGURE 1. Energy Level Diagram for ArI $(4p - 4s \left[\frac{3}{2}\right]^{\circ}$ transitions)



II. THEORY

Argon may be excited by proton bombardment by direct excitation:

$$H^{+} + Ar \rightarrow H^{+} + Ar^{*} \tag{1}$$

by charge transfer:

$$H^{\dagger} + Ar \rightarrow H + Ar^{\dagger} \tag{2}$$

and by simultaneous excitation and ionization:

$$H^{+} + Ar \rightarrow H^{+} + Ar^{+*} + e$$
 (3)

Subsequent to the proton bombardment, argon may reach the ith state by cascade from a higher state k:

$$Ar_k^* \rightarrow Ar_i^* + energy$$
 (4)

or by electron capture from the ionized state:

$$Ar^{+} + \bar{e} \rightarrow Ar_{i}^{*} + energy$$
 (5)

The observed spectral lines at 7635 Å and 8115 Å are both in the ArI spectrum so that only the direct excitation of equation (1), the cascade of equation (4), and the electron capture of equation (5) need be considered.

Once excited, the argon atoms will lose energy either by radiative transition to a lower state:

$$Ar^* \rightarrow Ar + hv$$
 (6)

or by collision with another argon atom:

$$Ar^* + Ar \rightarrow 2Ar + kinetic energy.$$
 (7)

If a proton beam of density n (ions/cc) and velocity v(cm/sec) is incident on the target gas of density N (atoms/cc), the rate of change



of the population of an excited state "i" of the target gas is expressed by Thomas and Bent [Ref. 6] as:

$$\frac{dN_{i}}{dt} = nv\sigma_{i}N + \sum_{k>i}A_{ki}N_{k} - \sum_{j
(8)$$

The first term gives the rate of population caused by direct excitation due to the incident proton beam in terms of the excitation cross section, $\sigma_{\bf i}$, for exciting to state i. The second term gives the rate of population of state i due to cascading from all states k higher than i in terms of the transition probability, $A_{\bf ki}$, for transition from k to i. The third term gives the rate of depopulation of state i to a lower state j in terms of the transition probability, $A_{\bf ij}$, for the transition from i to j. The fourth term represents secondary collision processes affecting the density of state i and has been examined more closely by Gabriel and Heddle [Ref. 7]. Atoms in state i can lose excitation by radiation or by collision. The rate of excitation loss can be expressed as:

$$-\frac{dN_{i}}{dt} = (A_{ij} + \sum_{j < i} A_{ij})N_{i} + \sum_{m} C_{im}N_{i}$$
(9)

 A_{il} is the transition probability from state i to the ground state and is zero for all states except transitions from the $4s\left[\frac{3}{2}\right]^r$ state. The first term is thus identical with the third term in equation (8) and represents the depopulation due to radiative transition. The second term in equation (9) represents the loss from state i due to collisions with other atoms in state m where C_{im} is the collision probability between states m and i and is given by:

$$C_{im} = 4N\sigma_{im} \left(\frac{kT}{\pi M}\right)^{1/2}$$
 (10)



where σ_{im} is the collisional de-excitation cross section and M is the atomic weight in grams.

One additional term is needed to represent the population of state i by electron capture. This term would be dependent on the population of the ionized state, N_r^+ , and the electron capture cross section, σ_{ri} , for capture into state i. Thus, a term for equation (5) would be represented by:

$$\sum_{r} \sigma_{ri} N_{r}^{\dagger} . \tag{11}$$

If one lets $K_{im} = C_{im}/N$ and assumes an equilibrium condition $(dN_i/dt = 0)$, equation (8) becomes:

$$nv\sigma_{i}N + \sum_{k>i}A_{ki}N_{k} + \sum_{r}\sigma_{ri}N_{r}^{+} = \sum_{j (12)$$

In order to get equation (12) into a usable form, the following assumptions were made:

- 1) P = NkT The perfect gas law is the equation of state for the target gas.
- 2) $\Sigma A_{ki}N_{k} = bA_{k}N$ The population of state k is proportional to the density of the target gas and b is constant.
- 3) $\Sigma \sigma_{ri} N_r^+ = c \sigma_r N \dots$ The population of the ionized state is proportional to the density of the target gas and c is constant.
- 4) $N_i = d\underline{I}$ The population of the ith state is proportional to the intensity (\underline{I}).
- 5) $\sum_{j < i} A_{ij} = A_i$ A constant for the line in question.
- 6) $\sum_{m} K_{im} = K_{i}$ A constant for the line in question.
- 7) $nv\sigma_i = a$ A constant for the line in question.



It has been shown in many thermodynamics textbooks [Ref. 8 and 9] that the ideal gas law is valid to within a few percent at pressures well above the 800 torr range of this experiment.

Substituting into equation (12) and performing all summations, one finds that:

$$\frac{aP}{kT} + \frac{bA_kP}{kT} + \frac{c\sigma_rP}{kT} = dA_i\underline{I} + \frac{dK_i}{kT}\underline{IP}.$$
 (13)

After rearranging terms and consolidating constants, this becomes:

$$P = A\underline{I} + BP\underline{I} \tag{14}$$

where

$$A = \frac{dkTA_{i}}{a + bA_{k} + c\sigma_{r}}$$
 (15a)

and

$$B = \frac{dK_1}{a + bA_k + c\sigma_r}$$
 (15b)

Dividing by I:

$$\frac{P}{I} = A + BP. \tag{16}$$

Thus, by plotting experimental data as P/\underline{I} versus P, a straight line with intercept A and slope B should result.

If one takes the ratio of B/A, the value of K_i (the collisional de-excitation rate) or A_i (the total transition probability from state i) can be determined:

$$K_{i} = \frac{B}{A} kTA_{i}. \tag{17}$$

A dimensional analysis of equation (14) yields the units of B/A to be $torr^{-1}$ which can be converted to cm^3 erg⁻¹ and is consistent with equation (17) in which K_i is in cm^3 sec⁻¹ from equation (10). Since there is one equation with two unknowns, K_i and A_i , it was decided to calculate the collisional de-excitation rate from equation (17). Thus, the value of



 A_i had to be found from the available literature. Wiese, Smith, and Miles [Ref. 10] have analyzed and tabulated the values of $\sum_{j< i} A_{ij}$ from many experiments. These values are comparable with the theoretically derived values of Garstang and Van Blerkom [Ref. 11] using jl coupling theory. Table I contains the comparisons.

Reference	Wavelength	$\sum_{j< i} A_{ij}$ (x 10 ⁸ sec)
(10)	7635	.380
(11)	7635	.351
(10)	8115	.366
(11)	8115	.381

TABLE I. Radiative Transition Probabilities

The experimentally obtained values of Wiese, et. al., were the ones used to calculate K_i as it was felt that they were more accurate.

The collisional de-excitation cross section, $\sigma_d = \sum_{m} \sigma_{im}$, can then be determined from equation (10):

$$\sigma_{d} = K_{i} \left(\frac{16 \text{ kT}}{\pi \text{M}} \right)^{-1/2} . \tag{18}$$

The value of T (temperature) was taken to be that of room temperature, 300 K.

Using equations (17) and (18) the collisional de-excitation rate, K_i , and the collisional de-excitation cross section, σ_d , were calculated for the two spectral lines studied in argon. The results are tabulated in Table II.



III. EXPERIMENTAL PROCEDURE

The experiment was performed using a High Voltage Engineering Corporation Model AK-N Van De Graaff Positive Ion Accelerator capable of energies up to 2.5 MeV. The Van De Graaff was used to produce protons which were accelerated into a target chamber where the argon gas pressure could be varied from about 10^{-6} to 800 torr. An aluminum foil window, approximately 1.6×10^{-3} centimeters thick, was placed between the target chamber and the accelerator drift tube in order to keep the accelerator components and drift tube under the 10^{-5} - 10^{-6} torr necessary for efficient operation. Hydrogen gas was used for the proton production and a magnetic analyzer in the drift tube insured that only H^+ particles were received in the target chamber. The aluminum foil window caused an energy loss in the proton beam of $0.25 \pm .05$ MeV [Ref. 12]. This window allowed the target gas pressure to be varied independently of the vacuum in the Van DeGraaff.

The target gas was fed, via a high pressure regulator, needle valve, and manifold system, into a pyrex collision chamber, as shown in Fig. 2.

The experimental apparatus consisted of a vacuum system, pressure measuring system, and the optical system. The vacuum system was composed of an oil diffusion pump and an independent fore pump and was capable of evacuating the target chamber to about 2×10^{-6} torr. Pressure measuring was accomplished by means of an ion gauge, thermocouple, and two Wallace and Tiernan pressure gauges. One gauge read from 0 to 50 torr and the second from 0 to 800 torr. The beam was viewed at 90° to the collision chamber by the optical apparatus.



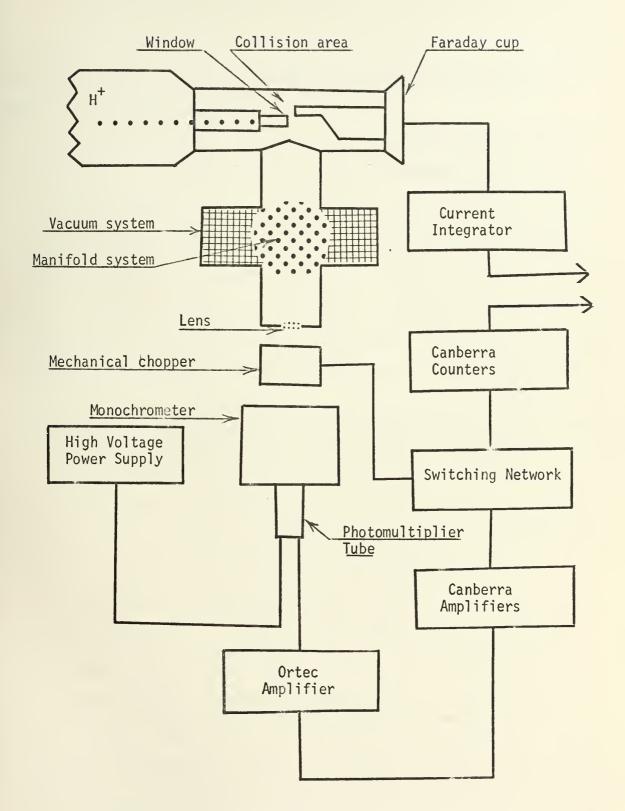


FIGURE 2. Schematic Diagram of Collision Chamber and Electronics



The target gas was excited by a proton beam of about one microampere and energy of about 1.5 MeV. The beam current was collected by a faraday cup and the total beam charge was measured by an Eldorado Electronics Model CI-110 current integrator.

The intensity of the excited argon gas was measured at 90° from the collision area. The light was focused by a 15 centimeter focal length quartz lens, through a mechanical chopper, and into the slits of a Jarral-Ash monochrometer with 500 micron slits. The slits were positioned perpendicular to the collision area so that only light from the first centimeter in front of the aluminum foil window was received. The energy loss in this portion of the beam was small, especially when compared to the energy loss that would have been measured if the slits had been placed parallel to the beam path. The resolving power of the monochrometer was \pm 10 Å about the wavelength being measured. One other transition for each line being measured (at 7628 Å and 8105 Å) was within this limit. Tests using an argon geisler tube and 100 micron slits with a resolving power of \pm 5 Å showed that the intensity of these lines was insignificant compared to the intensities of the 7635 Å and 8115 Å lines.

The light from the monochrometer passed into an RCA type 7102 photomultiplier tube housed in a dry ice cooled Electro-Optics Type PM-101 tube assembly and operated at a negative 1200 volts. The signal from the photomultiplier tube was fed into two Canberra 800 Series Amplifier-Counter systems. The counters were gated by a switching network activated at 78 Hz by the mechanical chopper. One counter was used to measure the signal from the light beam plus background noise. The second counter measured the background signal only.



Using the apparatus described above, intensity (\underline{I}) versus pressure (P) data was recorded. Since the intensity is proportional to the photomultiplier tube output registered on the counters, N_i , the intensity was calculated as:

$$\underline{I} = \frac{N_{C} - N_{b}}{QB}$$
 (19)

where $N_{\rm C}$ was the total number of counts, $N_{\rm b}$ the background count (both corrected for dead time), and QB the total proton beam charge collected. The value of QB was constant for each run in order to provide an equivalent excitation base for all experimental runs. Hence, \underline{I} is a relative intensity in arbitrary units. The pressure of the target gas was varied from 1 to 800 torr on each experimental run and \underline{I} was measured for each pressure considered. An IBM 360 computer was used to perform the necessary calculations and plot intensity versus pressure curves from the experimental data.



IV. RESULTS AND CONCLUSIONS

A. PRESSURE DEPENDENCE

Intensity versus pressure was measured for the 7635.1Å and 8115.3Å transitions in the ArI spectrum. A plot of intensity versus pressure for these lines is shown in figures 3 and 4.

Both transitions demonstrate that there is a functional dependence of the intensity on the pressure. This dependence rises rapidly up to about 150 torr where a leveling off effect occurs. This is followed by a general rise in the intensity up to the maximum pressures indicated. A comparison of the results of figures 3 and 4 with those of Tullington's work on helium [Ref. 13] and Smelley's work on N_2 [Ref. 12] indicates that the intensity did not rise nearly as quickly as the two previous studies. The same leveling off did occur, however, although not to the same extent.

A plot of P/I versus P, using the same data, shows a reasonable verification of equation (16), as shown in figures 5 and 6. The deviation from linearity can be shown to be within the deviations expected from using a digital counter. Attempts to reduce this scatter were made but to no avail. The signal to noise ratio of the argon lines, even at high pressures, was too low for good statistical data. At pressures less than about 50 torr, the background count was approximately 80% to 90% of the photon plus background count; while at pressures above 400 torr, it was about 20%. In fact, a series of consecutive counts at the same pressure and beam current settings produced data that varied up to 30%. Monochrometer slits of approximately 800 micron width were tried but the resolution was inadequate. A dove prism was also used during some experimental runs



which rotated the light beam 90° so that the entire collision area was parallel to and entering the slit. This technique reduced the energy resolution to an unacceptable level. Prior to the use of the counter apparatus described above, a second current integrator was used to measure the light intensity. The signal was passed through the mechanical chopper and into a lock-in amplifier where the light signal was converted to an AC signal fed into the current integrator. Experimental runs using this apparatus showed much less scatter of data points at the mid and high pressure end of the curve (Fig. 11 and 12). A breakdown of the current integrator forced the use of the counter apparatus.

On approximately one-half of the data sets a high initial P/I value was found which dropped sharply down to about 50 torr and then rose linearly as pressure was increased. An attempt was made to fit a theoretical curve to this data. In a few cases a good fit was obtained using a decreasing expotential function of the form:

$$\frac{P}{I} \propto -Ce^{-DP}$$
 (20)

as an additional term in equation (14). This could possibly represent excitation of the argon by high energy & rays produced from a previous collision of a proton with an argon atom. The random appearance and non-reproducibility of this phenomena caused it to be dropped from the theory. It is now believed that this is primarily attributable to the low signal-to-noise ratio at pressures below about 50 torr and is thus data point scatter. These points were omitted for the least square curve fitting which led to the theoretical curves shown in figures 7, 8, 9, and 10.



Wavelength (Å)	Ratio = B/A (torr ⁻¹)	K _i (cm ³ⁱ sec ⁻¹)	d (cm ²)
7635.1	3.21 ± .59 (-3)	3.78 ± .70 (-12)	6.7 ± 1.2 (-17)
8115.3	3.35 ± .41 (-3)	3.81 ± .53 (-12)	6.76 ± .95 (-17)
3.21 + .59 (-3) =	$= 3.21 + .59 \times 10^{-3}$		

TABLE II. Ratio, Collisional De-excitation Rate and Collisional De-Excitation Cross Section



B. COLLISIONAL DE-EXCITATION RATES AND CROSS SECTIONS

The ratio B/A was calculated for all experimental runs. Since the data points represented a straight line fit in accordance with equation (16) to within experimental error, it was felt that equations (17) and (18) were valid representations of the collisional de-excitation rates and collisional de-excitation cross sections. The results are displayed in Table II. The numbers shown represent an average of nine experimental runs of the 7635 $\mathring{\rm A}$ line and six of the 8115 $\mathring{\rm A}$ line which were considered valid. The approximate equality of the two sets of data is consistent with the expectation from the theory since the transition probabilities (Table I) are nearly equal. This equality could also be inferred from the closeness in energy level for the two upper states radiating to the same final state as shown in figure 1.

C. CONCLUSIONS

The functional dependence of intensity on pressure expressed in equation (14) appears to be valid for the two argon transitions studied. Improvements in experimental technique may present a more accurate indication of this dependence, particularly at low pressures. The two weak links in the present system are the sensitivity of the photo-multiplier tube and the use of counters rather than a second current integrator. This author has not been able to locate locally a photomultiplier tube with greater sensitivity in the near infrared region that was studied. As this paper is being written, steps are being taken to locally build a solid state current integrator to replace the present counter system.

Further study on the remaining lines of the 2p to $1s_5$ series needs to be done to see if the collisional de-excitation rates and cross sections remain approximately equal.



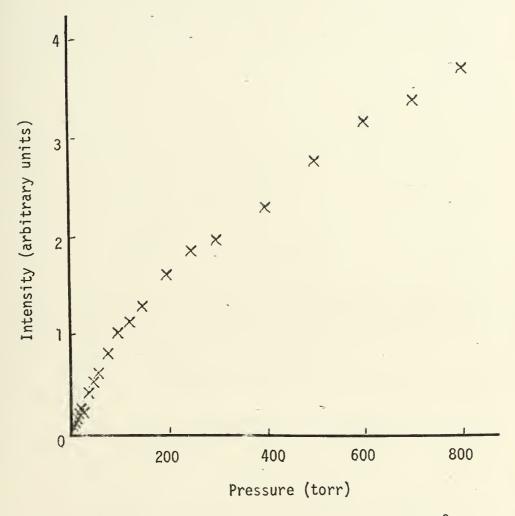


FIGURE 3. Intensity versus Pressure, 7635 Å



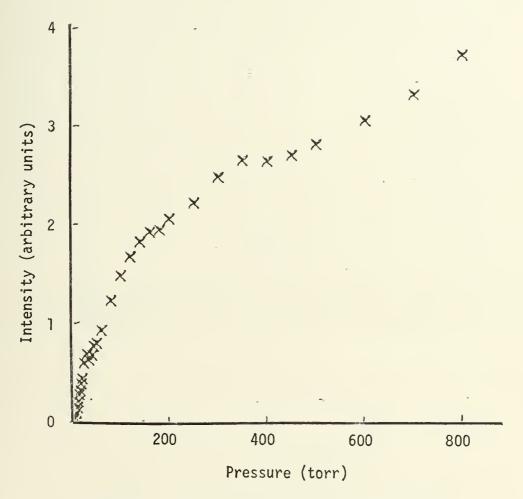


FIGURE 4. Intensity versus Pressure, 8115 Å



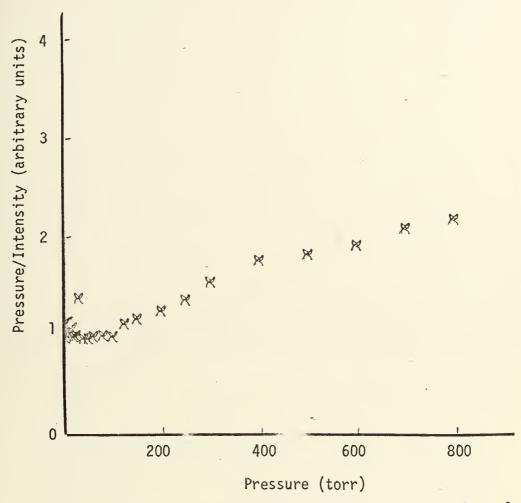


FIGURE 5. Pressure/Intensity versus Pressure, 7635 Å



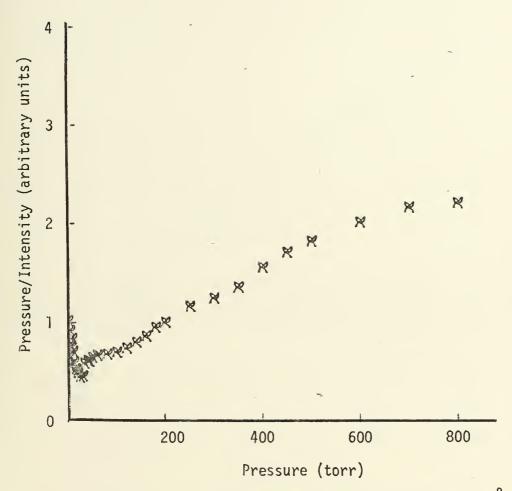


FIGURE 6. Pressure/Intensity versus Pressure, 8115 Å



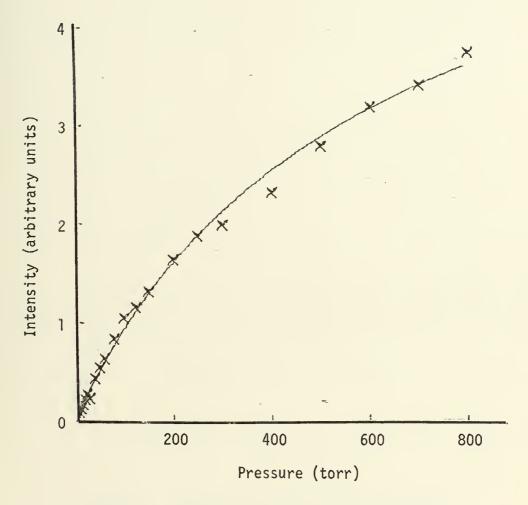


FIGURE 7. Intensity versus Pressure (Theory Comparison), 7635 Å



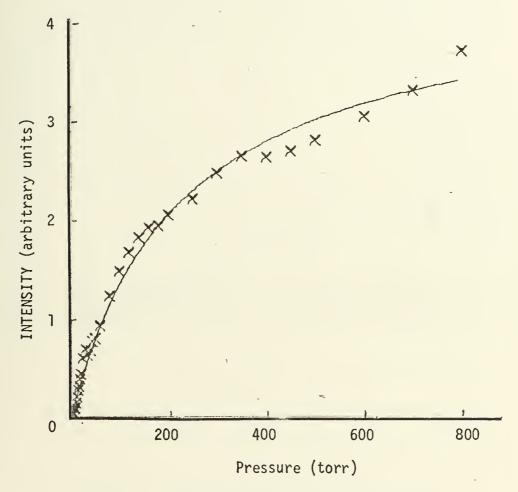


FIGURE 8. Intensity versus Pressure (Theory Comparison), 8115 Å



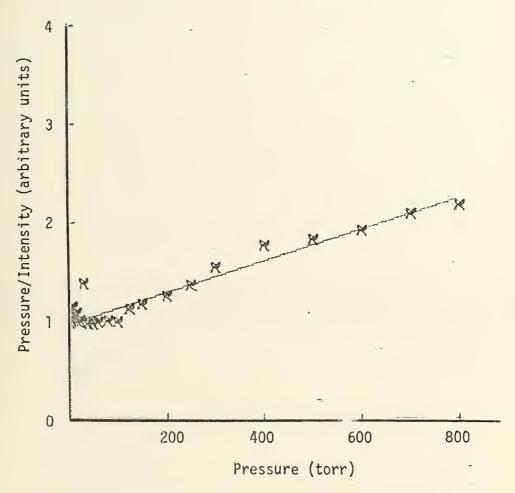


FIGURE 9. Pressure/Intensity versus Bressure (Theory Comparison), 7635 A



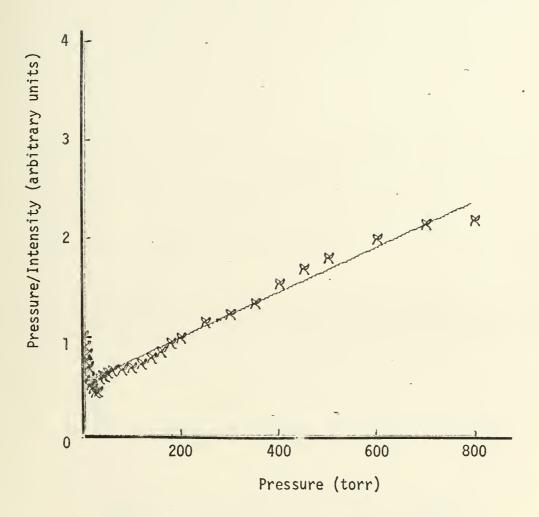


FIGURE 10. Pressure/Intensity versus Bressure (Theory Comparison), 8115 A



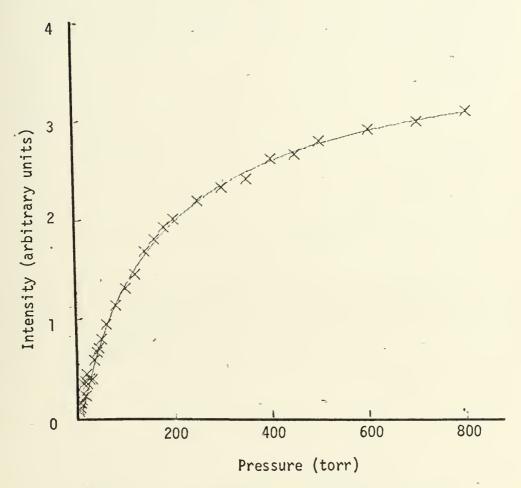


FIGURE 11. Intensity versus Pressure (Theory Comparison with data from two current integrator method), 7635 Å



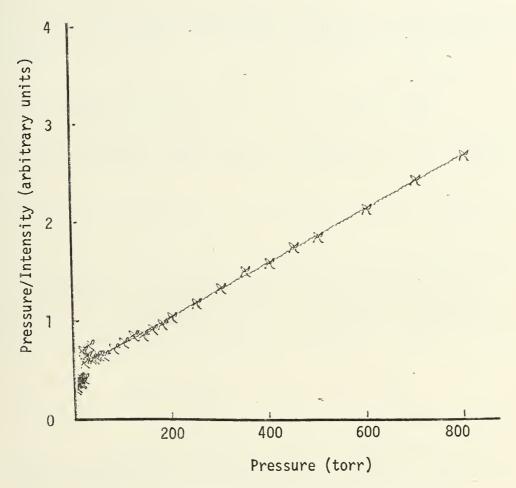


FIGURE 12. Pressure/Intensity versus Pressure (Theory comparison with data from two current integrator method), 7635 A



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ABSTRACT

Research grade argon at pressures from 1 to 800 torr was excited by 1.5 MeV protons from a Van De Graaff accelerator. The intensities of the 7635.1 Å and 8115.3 Å spectral lines were analyzed for their dependance on pressure. Theoretical equations were derived for the intensity as a function of pressure which agreed quite well with the experimental data. The collisional de-excitation rate, Kj, for the 7635 Å transition was found to be 3.78 \pm .70 x 10⁻¹² cm³ sec⁻¹ and the collisional de-excitation cross section, $\sigma_{\rm d}$, was found to be 6.7 \pm 1.2 x 10⁻¹⁷ cm². For the 8115 Å transition, Kj was found to be 3.81 \pm .53 x 10⁻¹² cm³ sec⁻¹ and $\sigma_{\rm d}$ was found to be 6.76 \pm .95 x 10⁻¹⁷ cm².



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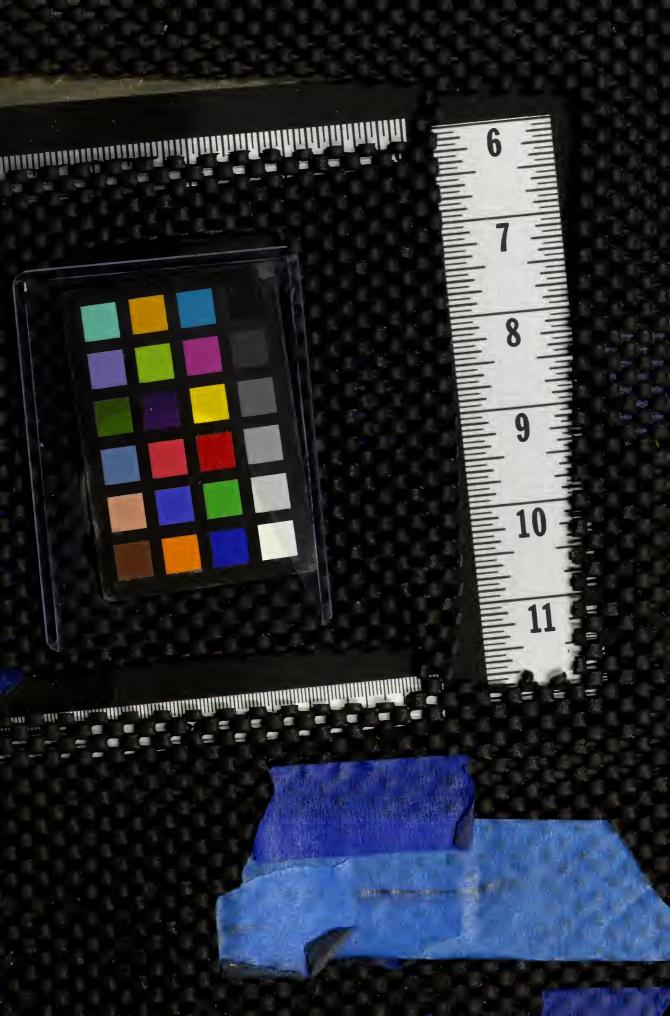






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